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Re:	U.S. Patent Application No. 10/054,213	Date:	Wednesday, January 12, 2005

☐ Confirmation will follow☒ No confirmation to follow

Examiner:

In response to your phone message, attached please find the referenced article from the amendment filed October 20, 2004.

If you have any further questions, please do not hesitate to contact us.

A. Froebrich

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Fuel Cell Research

Welcome to our page describing electrochemical instrumentation used in Fuel Cell research. Originally, PAR products were used by electrochemists to examine the initial science behind the operation of a fuel cell. Now, these same instrumentation systems are used everywhere from membrane development to final testing of the fuel cell or stacks of fuel cells to evaluate their long term performance.

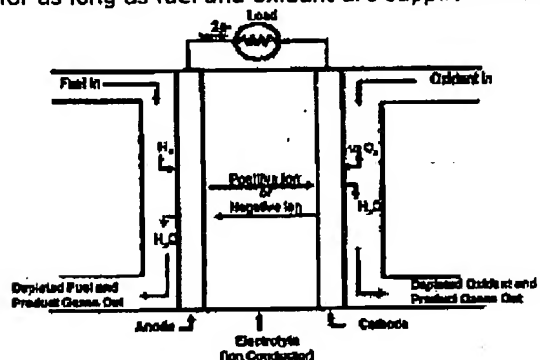
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What Is a Fuel Cell?

Fuel cells are electrochemical devices that convert the chemical energy of a reaction directly into electrical energy. The basic physical structure, or building block, of a fuel cell consists of an electrolyte layer in contact with a porous anode and cathode on either side.

The fuel cell is an energy conversion device that theoretically has the capability of producing electrical energy for as long as fuel and oxidant are supplied to the electrodes.



In a typical fuel cell, gaseous fuels are fed continuously to the anode (negative electrode) and an oxidant (i.e., oxygen from air) is fed continuously to the cathode (positive electrode); the electrochemical reactions take place at the electrodes to produce an electric current. A fuel cell, although having components and characteristics similar to those of a typical battery, differs in several respects. The battery is an energy storage device. The maximum energy available is determined by the amount of chemical reactant stored within the battery itself. The battery will cease to produce electrical energy when the chemical reactants are consumed (i.e., discharged). In a secondary battery, the reactants are regenerated by recharging, which involves putting energy into the battery from an external source. The fuel cell, on the other hand, is an energy conversion device that theoretically has the capability of producing electrical energy for as long as fuel and oxidant are supplied to the electrodes. Shown is a simplified diagram that demonstrates how the fuel cell works. In reality, degradation, primarily corrosion, or malfunction of components limits the practical operating life of fuel cells.

The electrolyte not only transports dissolved reactants to the electrode, but also conducts ionic charge between the electrodes and thereby completes the cell electric circuit, as illustrated in Figure 1-1. It also provides a physical barrier to prevent the fuel and oxidant gas streams from directly mixing. The functions of porous electrodes in fuel cells are: 1) to provide a surface site where gas/liquid ionization or de-ionization reactions can take place, 2) to conduct ions away from or into the three-phase interface once they are formed (so an electrode must be made of materials that have good electrical conductance), and 3) to provide a physical barrier that separates the bulk gas phase and the electrolyte. A corollary of item 1 is that, in order to increase the rates of reactions, the electrode material should be catalytic as well as conductive, porous rather

than solid. The catalytic function of electrodes is more important in lower temperature fuel cells and less so in high temperature fuel cells because ionization reaction rates increase with temperature. It is also a corollary that the porous electrodes must be permeable to both electrolyte and gases, but not such that the media can be easily "flooded" by the electrolyte or "dried" by the gases in a one-sided manner.

Electrochemistry of a Fuel Cell The ideal performance of a fuel cell is defined by its Nernst potential, E , or the ideal cell voltage. The overall reactions for various types of fuel cells are presented in the table below.

Fuel Cell	Anode Reaction	Cathode Reaction
Proton Exchange Membrane and Phosphoric Acid	$H_2 \rightarrow 2H^+ + 2e^-$	$\frac{1}{2} O_2 + 2H^+ + 2e^- \rightarrow H_2O$
Alkaline	$H_2 + 2(OH) \rightarrow 2H_2O + 2e^-$	$\frac{1}{2} O_2 + H_2O + 2e^- \rightarrow 2(OH)$
Molten Carbonate	$H_2 + CO_3^{2-} \rightarrow H_2O + CO_2 + 2e^-$ $CO + CO_3^{2-} \rightarrow 2CO_2 + 2e^-$	$\frac{1}{2} O_2 + CO_2 + 2e^- \rightarrow CO_3^{2-}$
Solid Oxide	$H_2 + O^{2-} \rightarrow H_2O + 2e^-$ $CO + O^{2-} \rightarrow CO_2 + 2e^-$ $CH_4 + 4O^{2-} \rightarrow 2H_2O + CO_2 + 8e^-$	$\frac{1}{2} O_2 + 2e^- \rightarrow O^{2-}$

CO - carbon monoxide

CO₂ - carbon dioxide

CO₃²⁻ - carbonate ion

e⁻ - electron

H⁺ - hydrogen ion

H₂ - hydrogen

H₂O - water

O₂ - oxygen

OH - hydroxyl ion

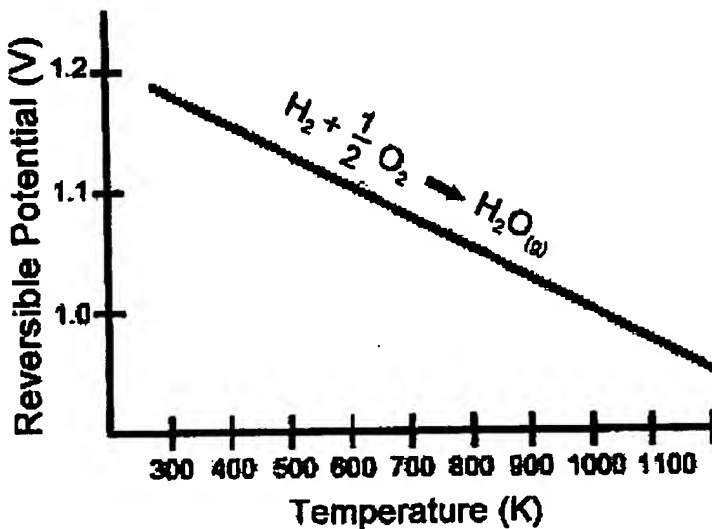
The corresponding Nernst equations for those reactions are provided in Table 2-2.

Cell Reactions*	Nernst Equation
$H_2 + \frac{1}{2} O_2 \rightarrow H_2O$	$E = E^0 + (RT/2F) \ln [P_{H_2}/P_{H_2O}] + (RT/2F) \ln [P_{O_2}]$

The Nernst equation provides a relationship between the ideal standard potential (E^0) for the cell reaction and the ideal equilibrium potential (E) at other temperatures and partial pressures of reactants and products. Once the ideal potential at standard conditions is known, the ideal voltage can be determined at other temperatures and pressures through the use of these Nernst equations. According to the Nernst equation for the hydrogen reaction, the ideal cell potential at a given temperature can be increased by operating at higher reactant pressures, and improvements in fuel cell performance have, in fact, been observed at higher pressures. The reaction of H₂ and O₂ produces H₂O. When a carbon-containing fuel is involved in the anode reaction, CO₂ is also produced. For MCFCs, CO₂ is required in the cathode reaction to maintain an invariant carbonate concentration in the electrolyte. Because CO is produced at the anode and consumed at the cathode in MCFCs, and because the concentrations in the anode and cathode feed streams are not necessarily equal, the CO₂ partial pressures for both electrode reactions are present in the second Nernst equation shown in Table 2-2.

The Ideal Performance of a Fuel Cell

The ideal standard potential (E^0) for a fuel cell in which H₂ and O₂ react is 1.229 volts with liquid water product, or 1.18 volts with gaseous water product. This value is shown in numerous chemistry texts (1) as the oxidation potential of H₂. The potential is the change in Gibbs free energy resulting from the reaction between hydrogen and oxygen. The difference between 1.229 volts and 1.18 volts represents the latent heat of vaporization of water at standard conditions.

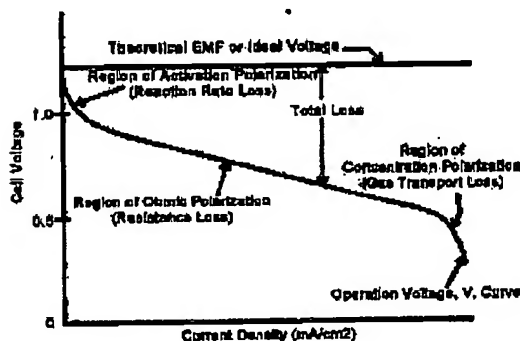


The figure below shows the relation of E to cell temperature. Because the figure shows the potential of higher temperature cells, the ideal potential corresponds to a reaction where the water product is in a gaseous state. Hence, E is 1.18 volts at standard conditions when considering gaseous water product. The impact of temperature on the ideal voltage, E , for the oxidation of hydrogen is also shown in types of fuel cells. Each case assumes gaseous products as its basis.

The ideal performance of a fuel cell depends on the electrochemical reactions that occur between different fuels and oxygen as summarized in Table 2-1. Low-temperature fuel cells (PEFC, AFC, and PAFC) require noble metal electrocatalysts to achieve practical reaction rates at the anode and cathode, and H_2 is the only acceptable fuel. With high-temperature fuel cells (MCFC and SOFC), the requirements for catalysis are relaxed, and the number of potential fuels expands. While carbon monoxide "poisons" a noble metal anode catalyst such as platinum (Pt) in low temperature fuel cells, it competes with H_2 as a reactant in high-temperature fuel cells where non-noble metal catalysts such as nickel (Ni) can be used.

Actual Performance of a Fuel Cell

The actual cell potential is decreased from its equilibrium potential because of irreversible losses, as shown below. Multiple phenomena contribute to irreversible losses in an actual fuel cell. The losses, which are called polarization, overpotential, or overvoltage, originate primarily from three sources: 1) activation polarization, 2) ohmic polarization, and 3) concentration polarization. These losses result in a cell voltage (V) that is less than its ideal potential, E ($V = E - \text{Losses}$).



The activation polarization loss is dominant at low current density. At this point, electronic barriers must be

overcome prior to current and ion flow. Activation losses increase as current increases. Ohmic polarization (loss) varies directly with current, increasing over the entire range of current because cell resistance remains essentially constant. Gas transport losses occur over the entire range of current density, but these losses become prominent at high limiting currents where it becomes difficult to provide enough reactant flow to the cell reaction sites.

Activation Polarization: Activation polarization is present when the rate of an electrochemical reaction at an electrode surface is controlled by sluggish electrode kinetics. In other words, activation polarization is directly related to the rates of electrochemical reactions. There is a close similarity between electrochemical and chemical reactions in that both involve an activation energy that must be overcome by the reacting species. In the case of an electrochemical reaction with $E_{act} > 50-100$ mV, it is customary to express the voltage drop due to activation polarization by a semi-empirical equation, called the Tafel equation.

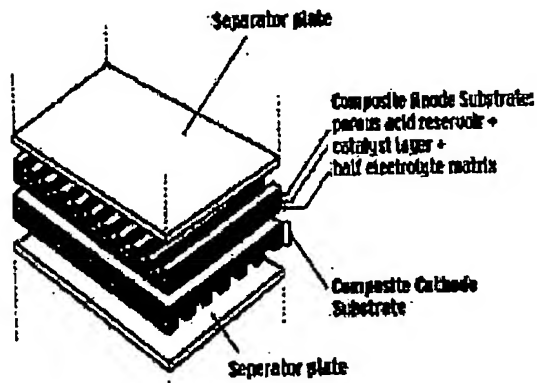
Tafel plots provide a visual understanding of the activation polarization of a fuel cell. They are used to measure the exchange current density, given by the extrapolated intercept at $E_{act} = 0$ which is a measure of the maximum current that can be extracted at negligible polarization, and the transfer coefficient (from the slope).

Ohmic Polarization: Ohmic losses occur because of resistance to the flow of ions in the electrolyte and resistance to flow of electrons through the electrode. The dominant ohmic losses through the electrolyte are reduced by decreasing the electrode separation and enhancing the ionic conductivity of the electrolyte. Because both the electrolyte and fuel cell electrodes obey Ohm's law, the ohmic losses can be expressed by the equation $E_{ohm} = IR$ where I is the current flowing through the cell, and R is the total cell resistance, which includes electronic, ionic, and contact resistance.

Concentration Polarization: As a reactant is consumed at the electrode by electrochemical reaction, there is a loss of potential due to the inability of the surrounding material to maintain the initial concentration of the bulk fluid. That is, a concentration gradient is formed. Several processes may contribute to concentration polarization: slow diffusion in the gas phase in the electrode pores, solution/dissolution of reactants and products into and out of the electrolyte, or diffusion of reactants and products through the electrolyte to and from the electrochemical reaction site. At practical current densities, slow transport of reactants and products to and from the electrochemical reaction site is a major contributor to concentration polarization.

Single Cells versus Stacked Cells

The figure below depicts a PAFC. As with batteries, individual fuel cells must be combined to produce appreciable voltage levels and so are joined by interconnects. Because of the configuration of a flat plate cell, see figure below, the interconnect becomes a separator plate with two functions: 1) to provide an electrical series connection between adjacent cells, specifically for flat plate cells, and 2) to provide a gas barrier that separates the fuel and oxidant of adjacent cells. The interconnect of a solid oxide fuel cell is a special case. All interconnects must be an electrical conductor and impermeable to gases. Other important parts of the cell are 1) the structure for distributing the reactant gases across the electrode surface and which serves as mechanical support, shown as ribs in figure, 2) electrolyte reservoirs for liquid electrolyte cells to replenish electrolyte lost over life, and 3) current collectors (not shown) that provide a path for the current between the electrodes and the separator of flat plate cells. Other arrangements of gas flow and current flow can be used in fuel cell stack designs.



Types of Fuel Cells

Alkaline Fuel Cell (AFC): The electrolyte in this fuel cell is concentrated (85 wt%) KOH in fuel cells operated at high temperature ($\sim 250^{\circ}\text{C}$), or less concentrated (35-50 wt%) KOH for lower temperature ($< 120^{\circ}\text{C}$) operation. The electrolyte is retained in a matrix (usually asbestos), and a wide range of electrocatalysts can be used (e.g., Ni, Ag, metal oxides, spinels, and noble metals). The fuel supply is limited to non-reactive constituents except for hydrogen. CO is a poison, and CO_2 will react with the KOH to form K_2CO_3 , thus altering the electrolyte. Even the small amount of CO_2 in air is detrimental to the alkaline cell.

Phosphoric Acid Fuel Cell (PAFC): Phosphoric acid concentrated to 100% is used for the electrolyte in this fuel cell, which operates at 150 to 220°C . At lower temperatures, phosphoric acid is a poor ionic conductor, and CO poisoning of the Pt electrocatalyst in the anode becomes severe. The relative stability of concentrated phosphoric acid is high compared to other common acids; consequently the PAFC is capable of operating at the high end of the acid temperature range (100 to 220°C). In addition, the use of concentrated acid (100%) minimizes the water vapor pressure so water management in the cell is not difficult. The matrix universally used to retain the acid is silicon carbide (SiC), and the electrocatalyst in both the anode and cathode is Pt.

Molten Carbonate Fuel Cell (MCFC): The electrolyte in this fuel cell is usually a combination of alkali carbonates, which is retained in a ceramic matrix of LiAlO_2 . The fuel cell operates at 600 to 700°C where the alkali carbonates are molten.

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